

Ionization of highly excited hydrogen atoms by a circularly polarized microwave field

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Some quantum-mechanical results describing the ionization of initially highly excited hydrogen atoms by a strong, circularly polarized microwave field are presented. A simplified two-dimensional model of the atom is used. Discrepancies between various classical estimates for the low-frequency ionization threshold are resolved.

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The ionization of a hydrogen atom which is initially prepared in a highly excited state by a linearly polarized microwave field [linearly polarized microwave ionization (LPMI)] has been intensively studied for the last 20 years, since the early experiment of Bayfield and Koch [1]. This system has served as one of the two most prominent examples of "quantum chaos" in atomic physics (the other being the hydrogen atom in a strong static magnetic field [2]). The progress in the understanding of the physics of LPMI has been frequently reviewed [3–5].

By contrast, relatively little attention has been paid to the complementary problem of ionization by a circularly polarized microwave field (CPMI), although the first theoretical treatments of this problem we believe appeared more than ten years ago [6, 7]. Only recently, while LPMI has become quite well understood, has the more difficult case of CPMI begun to be treated more comprehensively [8–14], although nonperturbative studies have up till now been restricted to classical calculations.

The difficulty in studying CPMI is easy to understand. In LPMI the projection of the atomic angular momentum on the light polarization axis is conserved. Therefore, one deals with an effectively two-dimensional system coupled by a periodic time-dependent electromagnetic wave. Here a simpler one-dimensional atomic model has frequently [3–5] been used, making the quantum calculations much easier. CPMI requires, in principle, a fully three-dimensional treatment, which becomes a very challenging problem in quantum mechanics.

Most of the classical studies have been restricted to a simplified two-dimensional model of hydrogen. The motion is then restricted to the plane defined by the rotation of the vector of polarization. Even in this case quite contradictory predictions, concerning ionization threshold, especially for low frequencies, have been reached [8–10, 14]. Experimental results are at present available for alkaline-earth-metal atoms [8, 9] only, but data for hydrogen should be available shortly [13].

In this Rapid Communication we present quantum-

mechanical calculations for CPMI of the two-dimensional hydrogen. Our results show reasonable agreement with classical numerical predictions [14] when such an agreement is expected and we also explain the origin of earlier discrepancies in the classical predictions.

First let us consider the two-dimensional model of hydrogen without any external perturbation. Its Hamiltonian (in atomic units) is obtained from the standard three-dimensional case by suppressing the z dependence:

$$H_0 = \frac{p_x^2 + p_y^2}{2} - \frac{1}{\sqrt{x^2 + y^2}}. \quad (1)$$

The quantum energies follow the Rydberg formula $E_n = -1/2n_*^2$, where the effective principal quantum number $n_* = n - 1/2$, $n = 1, 2, \dots$ rather than n appears. The states may be characterized by two quantum numbers: (n, m) , where $m = -(n-1), \dots, (n-1)$ is the eigenvalue of the angular momentum operator $L_z = xp_y - yp_x$.

In the presence of a circularly polarized electromagnetic wave, the Hamiltonian of the system reads

$$H = H_0 - Ff(t)[x \cos(\omega t) + y \sin(\omega t)], \quad (2)$$

where $f(t)$ denotes the envelope of the light (microwave) pulse with unit maximum and F measures the amplitude of the pulse. The oscillatory time dependence may be conveniently removed by passing into the rotating frame [15, 16]

$$\begin{aligned} \tilde{x} &= x \cos(\omega t) + y \sin(\omega t), \\ \tilde{y} &= -x \sin(\omega t) + y \cos(\omega t). \end{aligned} \quad (3)$$

This is accomplished quantum mechanically by applying the unitary transformation $U = \exp(i\omega L_z t)$. In the new frame,

$$\tilde{H} = \tilde{H}_0 - \omega \tilde{L}_z = \frac{\tilde{p}_x^2 + \tilde{p}_y^2}{2} - \frac{1}{\sqrt{\tilde{x}^2 + \tilde{y}^2}} - Ff(t)\tilde{x} - \omega \tilde{L}_z, \quad (4)$$

and the energy of the eigenstate of \tilde{H}_0 (n, m) becomes

$$\tilde{E}_n = -1/2n_*^2 - mw. \quad (5)$$

In the following we shall drop the tilde sign and work in the rotating frame only.

The microwave (light) pulse $Ff(t)$ may ionize the atom for sufficiently large F . To find the ionization yield for a given pulse and initial state we use the approach developed for strong-field ground-state ionization [17] — which we call the “single-state approximation” (SSA). We assume that the time variation of the pulse envelope $f(t)$ is slow in comparison with the average (outside of resonances) atomic time scale. Then one may assume that only the single state that evolves from the initial state is populated. Its instantaneous ionization rate may be calculated at fixed values of amplitude F . For instantaneous field values near a resonance, the complex energies undergo avoided crossings. It is assumed [17] that the electron remains in a diabatically evolving state. This assumption is valid provided the avoided crossing gaps are small in comparison with the time derivative of the pulse envelope. This should be the case for the system studied, as its corresponding classical dynamics is near integrable for fields below the classical ionization threshold [11], at least for frequencies smaller than the classical atomic frequency scale. The behavior of the CPMI of hydrogen is, in this sense, different from the LPMI, where the classical motion is much more irregular.

For a given F and putting $f(t) = 1$, Eq. (4) becomes time independent. The corresponding Schrödinger equation is transformed to the semiparabolic basis defined as $u = \sqrt{r+x}$, $v = \sqrt{r-x}$, where $r = \sqrt{x^2+y^2}$. Using next the standard complex rotation approach [18] to the resulting generalized eigenvalue problem, the complex energy of the state evolving from the initial state is found. Twice its imaginary part taken with opposite sign yields directly the instantaneous ionization rate at F , $\Gamma(F)$. The total ionization probability P in the SSA is found to be

$$P = 1 - \exp\left[-\int_0^T \Gamma(Ff(t))dt\right]. \quad (6)$$

We have performed several such diagonalizations with small steps in F for different ω values. We have checked numerically that for ω less than the classical atomic frequency (the case considered here), avoided crossings encountered during the evolution of the state under investigation are very small (compare also [9]).

For direct comparison with classical simulations [14], we assume that initially the atom is in the circular state (13, 12). $n = 13$ was chosen to compromise between the attempted study of the ionization of Rydberg states and the sizes of matrices leading to converged energies. Although various pulse shapes and pulse durations have been studied [20], we present here only the results for a $T = 100$ ps pulse with an envelope $f(t) = \sin^2(\pi t/T)$. Figure 1 presents the ionization thresholds, defined as 10% ionization (triangles) and 90% ionization (squares). The frequency and the maximum pulse amplitude have been rescaled using the scaling, familiar from the LPMI

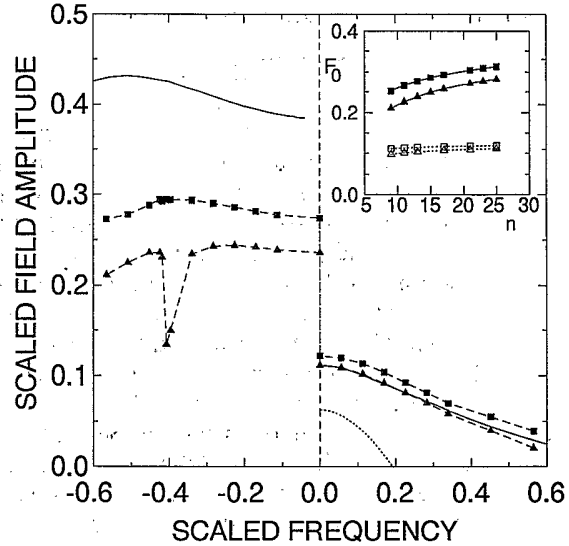


FIG. 1. Scaled amplitude of the pulse F_0 leading to the 90% ionization (squares) and to the 10% ionization (triangles) as a function of scaled frequency ω_0 . Connecting dashed lines are drawn to guide the eye. Vertical dashed line indicates static $\omega_0 = 0$ limit. Solid line for $\omega_0 > 0$ gives saddle-point classical prediction for the threshold obtained using (7); dotted line represents original prediction of [10]. Solid line for negative ω_0 gives the estimate of the breakup of periodic orbit stability and to a good accuracy reproduces the classical simulation result [14]. The inset shows the ionization-threshold dependence on the principal quantum number n , for scaled frequency $\omega_0 = 0.1$ (open squares, 90% ionization; open triangles, 10% ionization) and for $\omega_0 = -0.1$ (filled squares, 90% ionization; filled triangles, 10% ionization). For each n value the pulse duration has been appropriately scaled. Note the slow convergence of threshold values for negative ω_0 to the classical limit. For discussion see text.

case [3–5], in which the frequency of the atomic initial state is unity. For our two-dimensional hydrogen atom the proper scaling is $\omega_0 = n_*^3\omega$, $F_0 = n_*^4F$, where n_* is defined above.

The most striking feature of Fig. 1 is the apparent discontinuity at $\omega_0 = 0$; similar asymmetry between positive and negative frequencies has been observed in classical simulations [14]. To understand its origin, observe that Eq. (4) represents in the static limit ($\omega \rightarrow 0$) the well-known Lo Surdo–Stark effect. The eigenstates of Eq. (4) are then not (n, m) but (n, k) , and their energies are given in the perturbation limit by $E_{n,k} = -1/2n_*^2 + 3/2kn_*F$ with $k = -(n-1), \dots, (n+1)$. The states with maximal negative k are the lowest states of a given n_* manifold and lie close to the saddle of the potential. Their ionization threshold is given by $F_{\text{neg}} = 1/9n_*^4$ [8]. On the contrary, highest-energy states of the manifold, with maximal positive k , are located far from the saddle on the other side of the nucleus (in the region where the potential is maximal at negative x). They do not feel the influence of the saddle at all. As the Lo Surdo–Stark problem is separable, these states lie on well-defined tori, and they are not sensitive to whether their energy is lower or higher than the classical escape energy, given by the saddle energy.

For $\omega \neq 0$, the first-order perturbative result yields [19]

$$E_{n,k} = -1/2n_*^2 + k\sqrt{(3/2n_*F)^2 + \omega^2}, \quad (7)$$

which reduces to (5) for $F = 0$ under the $m = -k$ identification. Clearly for both ω and F small and positive and for maximal positive m (negative k), as is the case in Fig. 1, the initial state evolves into the state located close to the saddle (and its ionization threshold should be close to the static value of $F_0 = 1/9$), while for low negative ω it evolves into the state with maximal positive k and its ionization threshold should be much higher (compare Fig. 1). Due to the symmetry of (4) a change of $m \rightarrow -m$ is equivalent to the substitution $\omega \rightarrow -\omega$.

Strictly, one should point out that the SSA adopted here breaks down in the immediate vicinity of $\omega = 0$ when the pulse duration becomes comparable with the period of the electromagnetic wave. Thus the region $|\omega T| < 1$ is not well described by the present method. This is the region where the mixing of different m states forming the k state occurs just at the beginning of the pulse. For a $T = 100$ ps pulse and $n = 13$ it corresponds to the small interval $|\omega_0| < 0.005$, invisible on the scale of Fig. 1 but contributing to the apparent discontinuity at $\omega = 0$. One may use the SSA in the immediate vicinity of $\omega = 0$, but then as the initial state one should not use the m states but rather the k states. In this way *two* values for the threshold at $\omega = 0$ have been obtained. One corresponds to $k = -(n - 1)$ and represents the limit for positive ω , while the other one, for $k = (n - 1)$, is the limit for negative ω . To obtain the threshold for the circular initial state quantum mechanically, a fully-time-dependent approach would be necessary.

It is clear from the above that classical predictions based on the saddle-point criterion may give reasonable estimates for $\omega > 0$ only [with initial state $(n, m) = (n, n - 1)$]. Such a prediction has been obtained in [10]. Unfortunately, the author compared the energy of the saddle with the unperturbed energy $E = -1/2n_*^2$, which yields (see Fig. 1 dotted line) a wrong result [21]. Using the same approach, but utilizing (7) as the proper energy of the state (for details see [20]), the solid line in Fig. 1 ($\omega_0 > 0$ only) is obtained. This agrees quite well with quantum predictions, bearing in mind that the latter depend on the pulse duration (for long pulses the tunneling contributes, lowering the quantum prediction).

The quantum results agree quite well with classical simulations [14] for $\omega > 0$, while for $\omega < 0$, classical thresholds are significantly higher. This is to be expected following arguments similar to those already given. If, in the classical case, the field were switched slowly enough, one would follow the periodic orbit corresponding to the initial circular orbit. For long pulses, the classical threshold then corresponds to the field at which this orbit loses its stability—represented by the solid line for $\omega < -0.04$ in Fig. 1. For shorter pulses, when adiabaticity is lost, one may “lose” the periodic orbit earlier, while switching the field. The thresholds predicted by the stability breakup of the periodic orbit will differ strongly from the classical simulation results obtained with fixed pulse duration in the limit $\omega \rightarrow 0$, as the adiabaticity is then also

broken, due to the new time scale associated with ω^{-1} .

The quantum threshold is determined not by the breakup of the periodic orbit stability, but rather by the breakup of the torus corresponding to this state (which surrounds the periodic orbit and the transverse action of which is equal to $\hbar/2$). We have checked that, classically, the breakup of this torus causes the atom to ionize. The quantum threshold for $\omega_0 < 0$ does not scale classically for moderate n ; the smaller the \hbar (the higher the n), the “closer” the quantizable torus lies to the periodic orbit, and the higher will be the ionization threshold. This has been verified by studying ionization of different n states (see inset in Fig. 1; the corresponding classical threshold values can be easily read off from the main panel of the figure).

For very long pulses, the convergence of quantum and classical thresholds will be slow for $\omega_0 < 0$, while for short pulses (when classical calculations “lose” periodic orbits due to nonadiabaticity), classical-quantum correspondence should be established for lower initial n .

For positive ω_0 , when the threshold is determined by the saddle-point criterion, the quantum-classical correspondence is established for n as low as 10, as can be seen in the inset in Fig. 1.

Inspection of the quantum results in Fig. 1 reveals a resonant dip in the 10% ionization threshold around $\omega_0 = -0.4$. It has been verified that this dip is due to a partial enhancement of the ionization when the two-photon resonance $(n, m) \leftrightarrow (n + 1, m - 2)$ is encountered during the pulse rise. Near the corresponding (narrow) avoided crossing the instantaneous ionization rate becomes enhanced, leading to a temporal increase in the ionization yield. If the ionization threshold is defined at a low level (10%), this increase may lead to a lower threshold being obtained. A detailed study of this effect will be given elsewhere [20].

In summary, we have presented quantum predictions concerning ionization of an initially highly excited hydrogen atom by strong circularly polarized light. The results are in good agreement with classical simulations, when possible. The origin of the remaining discrepancies has been explained. The results, although obtained for a simplified two-dimensional atomic model, should be experimentally verifiable. It is now possible to create circular states that are well oriented in space; e.g., by the crossed fields method [22]. Choosing the plane of polarization vector rotation in the same plane should enable the results we have presented to be tested since for high n the circular states are approximately two-dimensional. It is quite important to generalize both the classical and the quantum results to a truly three-dimensional case. Such calculations are in progress and the preliminary classical results show that the asymmetry observed here is partially preserved for orbits crossing the polarization plane at a not too large angle. This strongly indicates that the effect should be found in the three-dimensional quantum calculations too. The two-dimensional model is, of course, unsuitable for a study of ionization from the ground state or the lowest excited states. There a fully three-dimensional calculation is necessary [23].

Finally, let us mention that the results presented here

are restricted mainly to the hydrogen atom or hydrogen-like ions. For other atoms the residual interaction with the core may significantly affect the ionization process [8, 9].

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